# Plasma Assisted Combustion Mechanism for Small Hydrocarbons

Andrey Starikovskiy Nickolay Aleksandrov



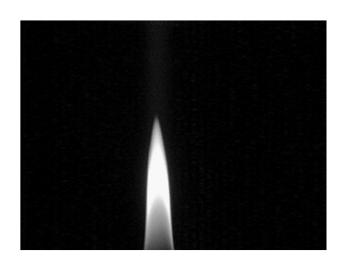


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**Report Documentation Page** 

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## Ignition, Combustion and Flame Control by Nonequilibrium Plasma

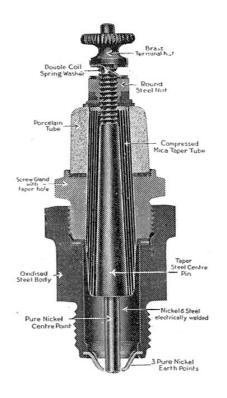


1814 - Brande. Flame/Field Interaction

W.T. Brande. Phil. Trans. Roy. Soc., 1814, 104, 51.

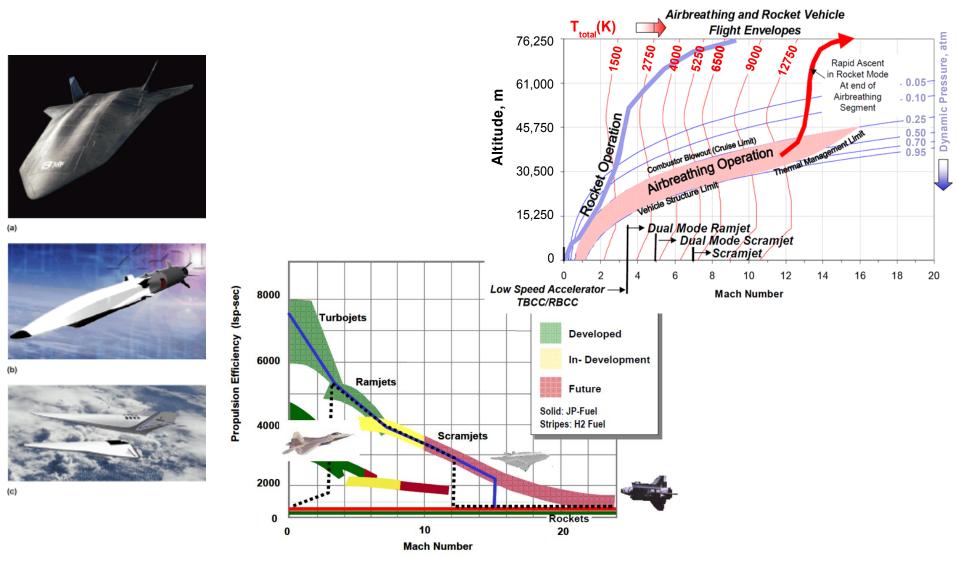


In 1860 Étienne Lenoir used an electric spark plug in his gas engine, the first internal combustion piston engine and is generally credited with the invention of the spark plug



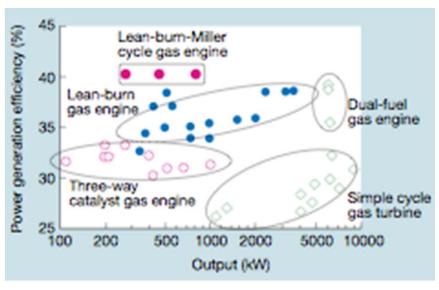
Physics of Nonequilibrium Systems Laboratory

### Propulsion Efficiency and Operating Regimes for Variety of Flight Systems

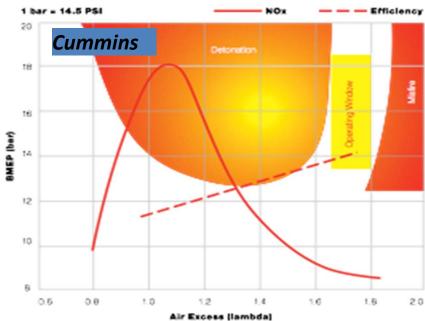


**Physics of Nonequilibrium Systems Laboratory** 

### **Lean Ignition for Gas IC Engines**



- Regular spark plugs  $\lambda < 1.4$
- Regular spark plugs
   with thin (Iridium/Platinum)
   electrodes λ < 1.6</li>
- RF, "plasma", etc. plugs  $\lambda < 1.8$









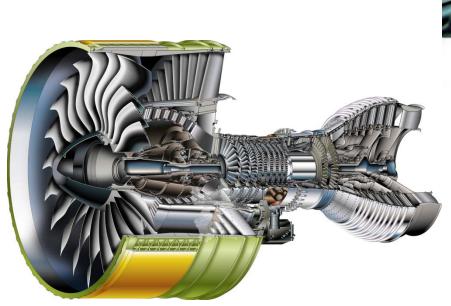


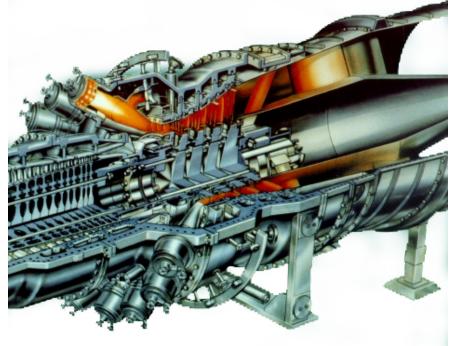
### **GTE Lean Regimes**

T = 700 - 1300 K

P = 20 - 30 atm

W = 10 - 1000 MW

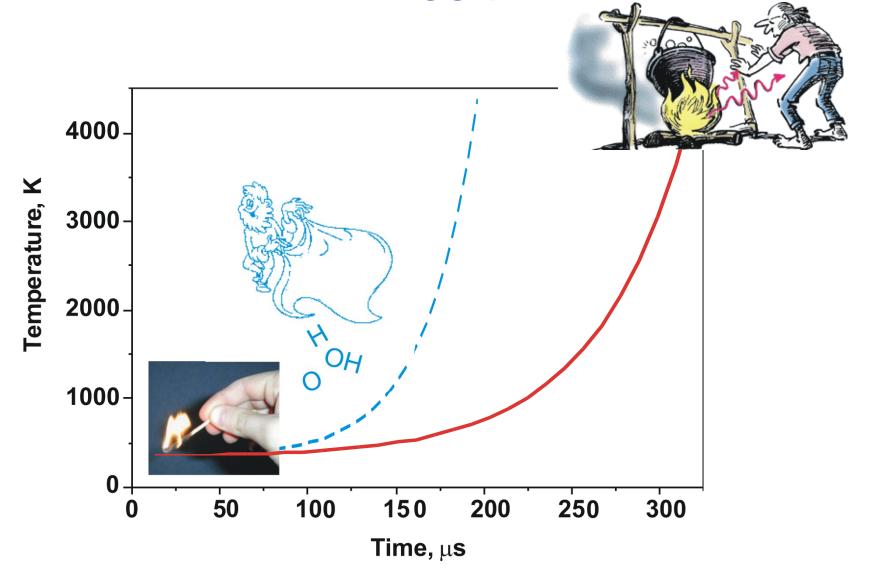






Physics of Nonequilibrium Systems Laboratory

Decreasing of Ignition Delay Time - 1994



## Kinetic Model: Previous Versions

**D.V.Zatsepin, S.M.Starikovskaia, A.Yu.Starikovskii** *Hydrogen oxidation in a stoichiometric hydrogen-air mixtures in the fast ionization wave*. Combust. Theory Modeling, 2001. V.5 pp.97-129.

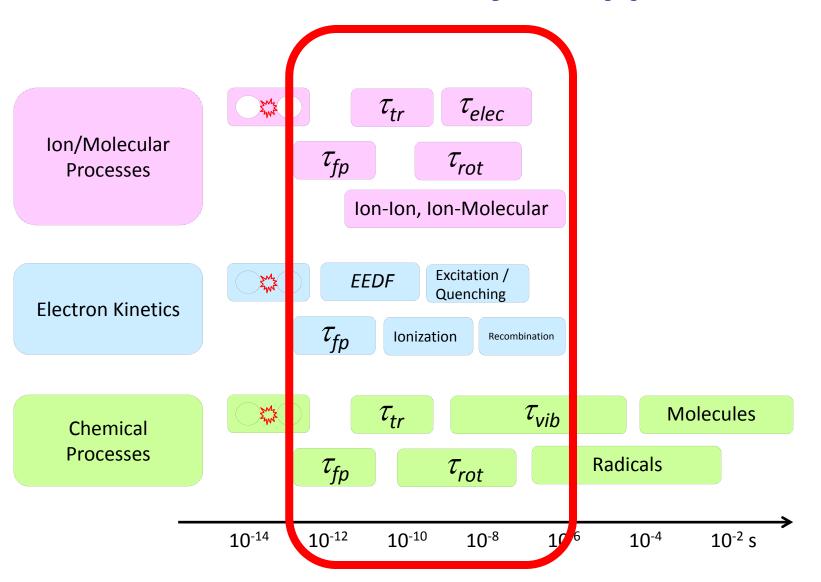
**N.A.Popov.** Effect of a Pulsed High-Current Discharge on Hydrogen—Air Mixtures. Plasma Physics Reports, 2008, Vol. 34, No. 5, pp. 376–391.

I.N. Kosarev, N.L. Aleksandrov, S.V. Kindysheva, S.M. Starikovskaia, A.Yu. Starikovskii. Kinetics of ignition of saturated hydrocarbons by nonequilibrium plasma: C2H6- to C5H12-containing mixtures. Combustion and Flame 156 (2009) 221–233

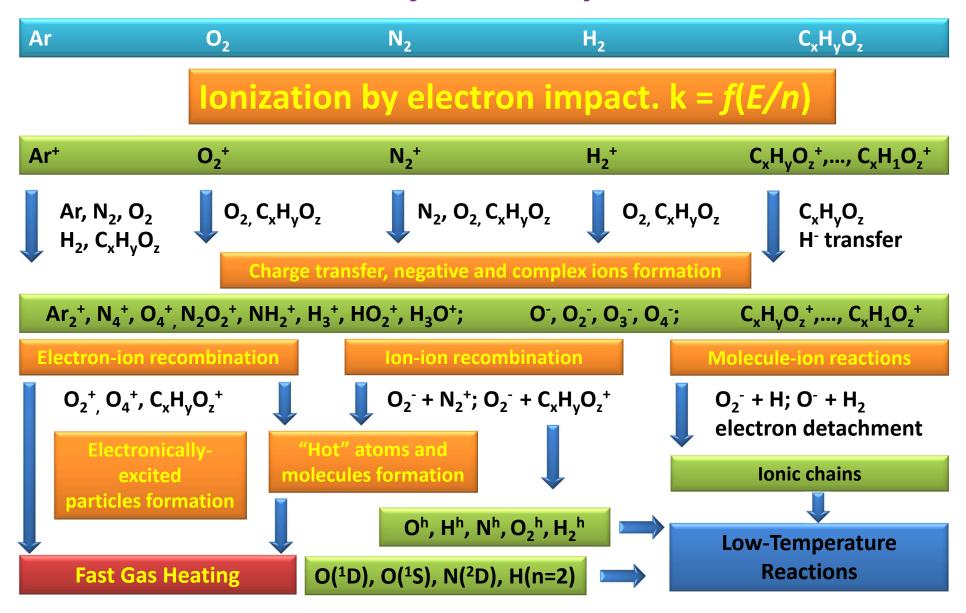
**A.Starikovskiy, N.Aleksandrov.** *Plasma-assisted ignition and combustion.* Progress in Energy and Combustion Science 39 (2013) 61-110

### **Predictive Modeling:**

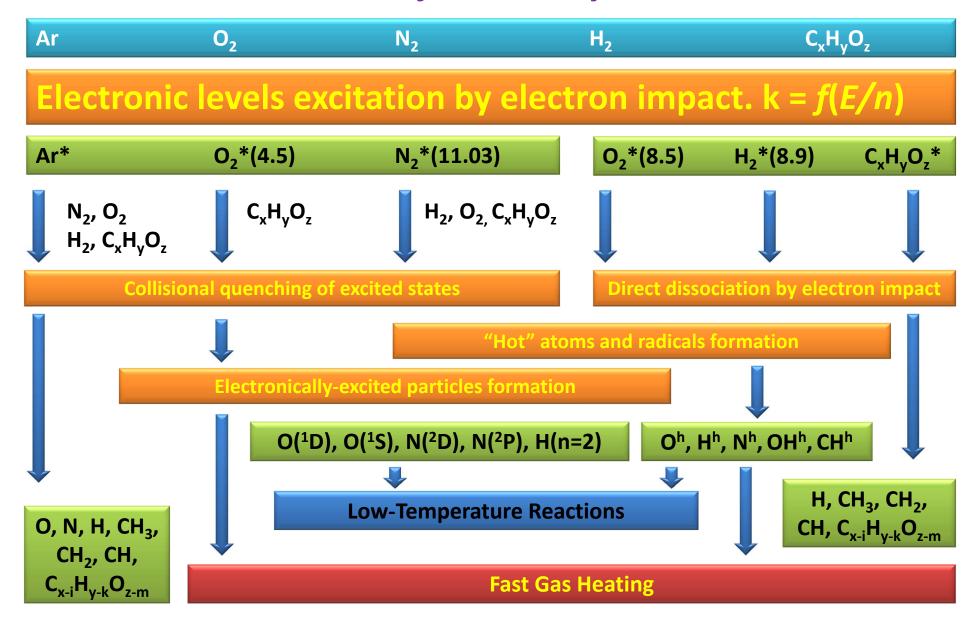
### **Key to Applications**



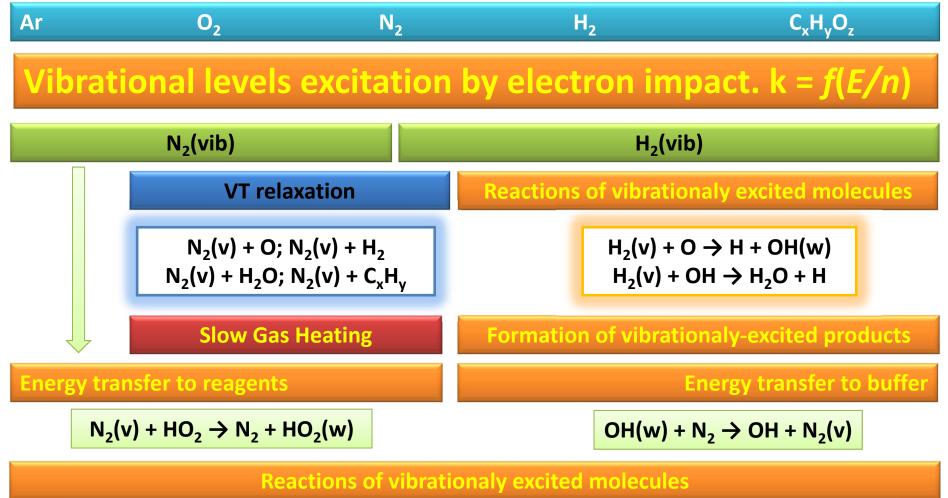
**Major Pathways** 



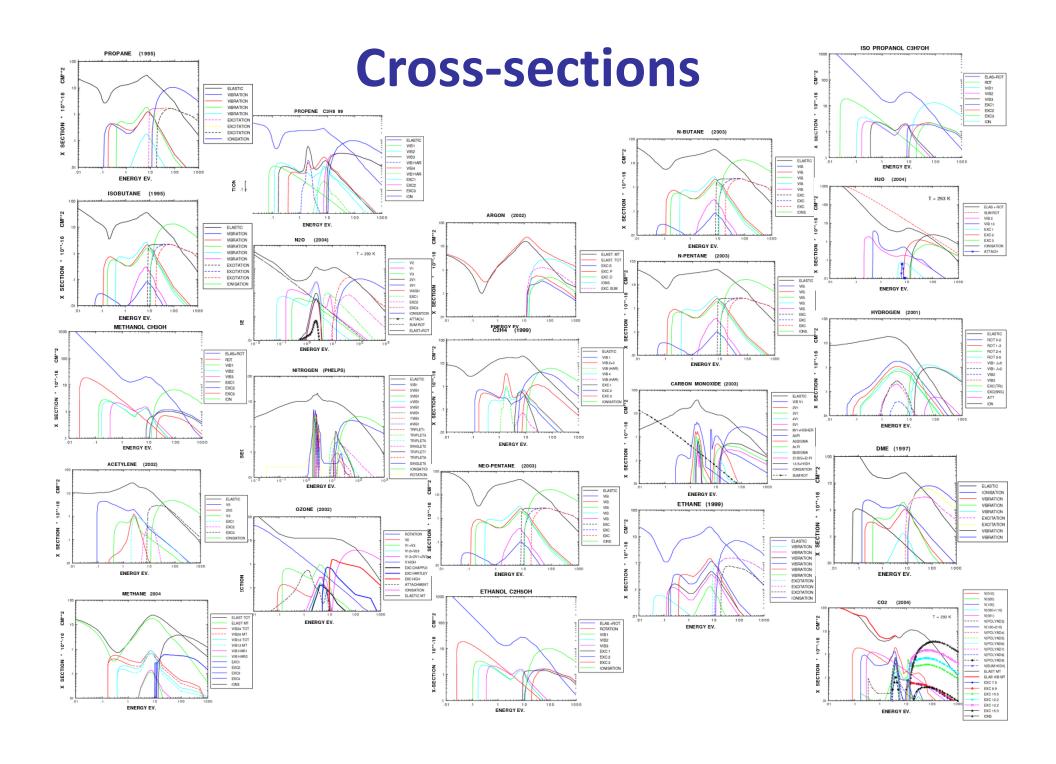
**Major Pathways** 



**Major Pathways** 



$$HO_2(w) \rightarrow H + O_2$$
  $OH(w) + H_2 \rightarrow H_2O + H$ 

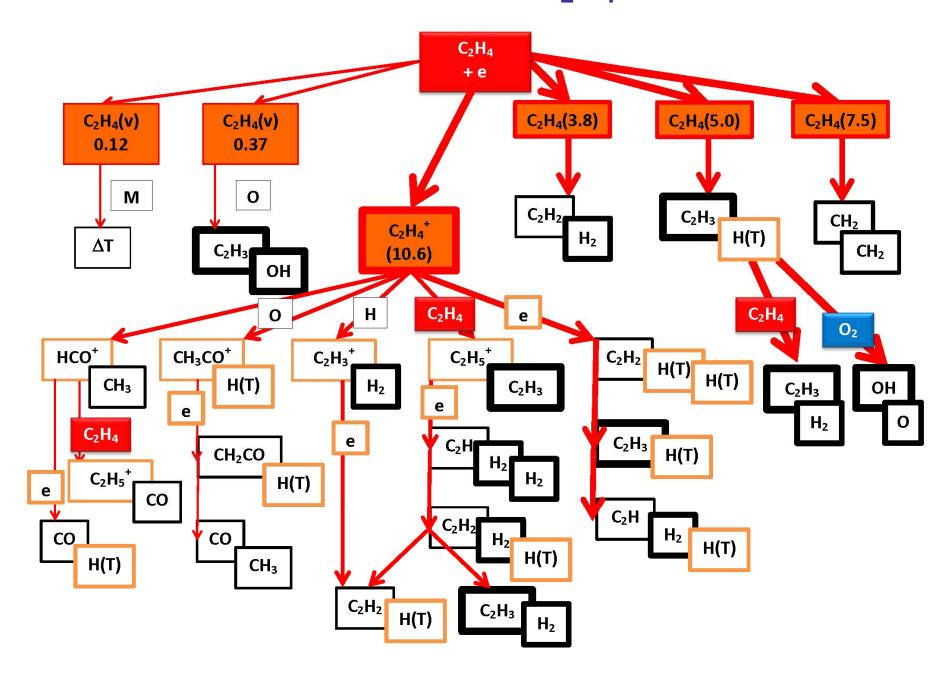


#### **Cross-sections Available**

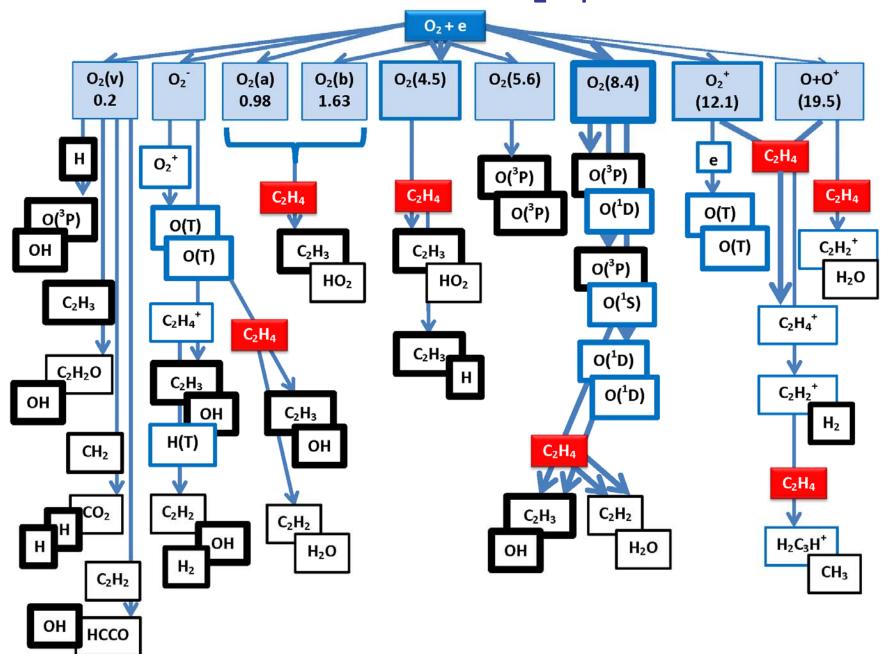
Atmospheric	Saturated	Unsaturated	Oxygenated	Isomers
N2	CH4	C2H2	СО	iso-butane
O2	C2H6	C2H4	СНЗОН	iso-propane
CO2	С3Н8	С3Н6	С2Н5ОН	neo-pentane
H2O	C4H10		CH3OCH3 DME	
О3	C5H12			
Ar	H2			
N2O				

#### **PAC Kinetic Mechanism** N2(+)+O2=N2+O2(+)N2(c3pu) => N2(b3pg)N2(c3pu) => N2(b3pg) + hnN2(+)+O2=>N2+O2(+)N2(c3pu)+N=>N+N2(b3pg)N2(+)+O2=>O2(+)+N2N2(c3pu)+N=>N(2p)+N2H2(v)+N2=H2+N2N2(+)+O=N+NO(+)LT /-144.9 0.0/ N2(c3pu)+N2=>N2+N2O2(v) + H = O + OH(v)O(-)+N(+)=N+ON2(c3pu)+N2=>N2(a'1)+N2N2(+)+O=N2+O(+)N2(c3pu)+N2=N2+N2(a'1su)H2(v) + O = H + OH(v)H2(v)+O2=H2+O2O(-)+N(+) +O2=>NO+O2LT /-144.9 0.0/ CH4(v) + O = CH3 + OH(v)N2(c3pu)+N2=N2+N2(a1su)N2(+)+O3=>N2+O+O2(+)O(-)+N(+)+N2=>N2+NOC2H2(v) + O = C2H + OH(v)N2(+)+N2(a3su)=>N+N3(+)H2(v)+CO2=H2+CO2C2H4(v) + O = C2H3 + OH(v)N2(c3pu)+N2=N2+N2(b3pg)N2(+)+NO=N2+NO(+)O(-)+O(+)=>O+O(5p)LT /-144.9 0.0/ C2H6(v) + O = C2H5 + OH(v)O(-)+O(+)=>O(1d)+O(5s)C2H5OH(v) + O = H2C2O + OH(v)N2(+)+N2O=>N2+N2O(+)O(-)+O(+)=O+ON2(c3pu)+O2=>N2+2O(T)H2(v)+H2O=H2+H2OC3H8(v) + O = C3H7 + OH(v)N2(c3pu)+O2=>N2+2O(T)N2(+)+N=N(+)+N2LT /-144.9 0.0/ C4H10(v) + O = C4H9 + OH(v)N2(c3pu)+02=>N2(a3su)+02O(-)+O(+)+O2=>O2+O2N2(c3pu)+O2=>N2+O(T)+O(1d)N2(+)+H2=H2(+)+N2C5H12(v) + O = C5H11 + OH(v)O(-)+O(+)+N2=>N2+O2H2(v)+H2=H2+H2N2(c3pu)+02=>N2+02 LT /-144.9 0.0/ N2(+)+H2O=H2O(+)+N2O(-)+N2(+)=>N2+OH2+OH(V)=H+H2ON2(c3pu)+N0=>N2+N0 O(-)+N2(+)=>N+N+OH2(v)+OH=H2+OH CH4+OH(V)=CH3+H2ON2(+)+CO => CO(+)+N2O(-)+N2(+)+N2=>N2+N2OLT /-144.9 0.0/ C2H4+OH(V)=C2H3+H2ON2(+)+CO2=>CO2(+)+N2O(-)+N2(+)+O2=>N2O+O2C2H2+OH(V)=CH3+CON2(c3pu)+H2=>N2+2H(T)H2(v)+H=H2+HC2H6+OH(V)=C2H5+H2ON2(+)+N2+N2=N2+N4(+)O(-)+O2(+)=>O+O2LT /0.0 0.0/ N2(c3pu)+CH4=>N2+CH3+H(T)C3H8+OH(V)=NC3H7+H2OO(-)+O2(+)=>O+O+ON2(c3pu)+CH4=>N2+CH2+2H(T)N2(+)+N2+M=>N4(+)+MO(-)+O2(+)+O2=>O2+O3C4H10+OH(V)=PC4H9+H2OH2(v)+O=H2+ON2(c3pu)+CH4=>N2+CH2+H2 NC5H12+OH(V)=C5H11-1+H2OO(-)+O2(+)+N2=>N2+O3LT /0.0 0.0/ N2(c3pu)+CH4=>N2+CH+H(T)+H2N2(+)+N+N2=>N2+N3(+)O(-)+N3(+) =>N+N2+ON2(c3pu)+C2H2=>N2+C2H+H(T)N2(+)+H2=>HN2(+)+HN2(c3pu)+C2H2=>N2+CH+CH O(-)+N4(+) =>N2+N2+OH(T) + H2 = H + H2O(-)+O4(+)=>O+O2+O2CH4(v)+CH4=CH4+CH4N2(c3pu)+C2H4=>N2+C2H3+H(T)H(T) + M = H + MN2(+)+CH4=CH2(+)+N2+H2LT /-40.0 0.0/ N2(c3pu)+C2H4=>N2+C2H2+2H(T)N2(+)+CH4=CH3(+)+N2+HO(T) + H2 = O + H2O(-)+H2(+)=>H+OHN2(c3pu)+C2H4=>N2+CH2+CH2 N2(+)+CH4=>N2+CH3(+)+HO(T) + M = O + MO(-)+CH4(+)=>H2+CH3CH4(v)+M=CH4+MO(-)+C2H2(+)=>H2+C2HLT /-61.0 0.0/ N2(c3pu)+C2H6=>N2+C2H5+H(T)N2(+)+C2H5OH=>N2+H5C2O(+)+HO(-)+C2H5OH(+)=>H2+H5C2OO2 + H(T) = O + OH(v)N2(c3pu)+C2H6=>N2+C2H4+2H(T)N2(+)+C2H2=>N2+C2H(+)+HN2(c3pu)+C2H6=>N2+C2H4+H2 O(-)+C2H4(+)=>H2+C2H3C2H6(v)+M=C2H6+MH2 + O(T) = H + OH(v)N2(c3pu)+C2H6=>N2+CH3+CH3 O(-)+C2H6(+)=>H2+C2H5LT /0.0 0.0/ CH4 + O(T) = CH3 + OH(v)C2H(+)+e=C2+HO(-)+C3H8(+)=>H2+C3H7C2H2 + O(T) = C2H + OH(v)N2(c3pu)+C2H5OH=>N2+C2H4OH+H(T)C2H(+)+e=CH+CO(-)+C4H10(+)=>H2+C4H9C2H2(v)+M=C2H2+MC2H4 + O(T) = C2H3 + OH(v)N2(c3pu)+C2H5OH=>N2+C2H3OH+2H(T)C2H(+)+C2H2=C4H2(+)+HO(-)+C5H12(+)=>H2+C5H11LT /0.0 0.0/ C2H6 + O(T) = C2H5 + OH(v)N2(c3pu)+C2H5OH=>N2+C2H5+HO C4H2(+)+C2H2=C6H3(+)+HN2(c3pu)+C2H5OH=>N2+CH3+CH2OH C2H5OH + O(T) = H2C2O + OH(v)C4H2(+)+C2H2=C6H4(+)+PHOTONC2H4(v)+M=C2H4+MC3H8 + O(T) = C3H7 + OH(v)C4H2(+)+e=C4H+HN2(c3pu)+C3H8=>N2+C3H7+H(T)LT /0.0 0.0/ C4H10 + O(T) = C4H9 + OH(v)N2(c3pu)+C3H8=>N2+C3H6+2H(T)C6H3(+)+e=C6H+H2O(-)+NO(+) =>NO+OC5H12 + O(T) = C5H11 + OH(v)N2(c3pu)+C3H8=>N2+C3H6+H2 C6H3(+)+e=C6H2+HO(-)+NO(+) =>N+O+OC2H5OH(v)+M=C2H5OH+M N2(c3pu)+C3H8=>N2+CH3+C2H5 C6H4(+)+e=C6H+H2+HO(-)+NO(+)+O2=>NO2+O2LT /0.0 0.0/ C6H4(+)+e=C6H2+H2O(-)+NO(+)+N2=>N2+NO2N2(c3pu)+C4H10=>N2+C4H9+H(T)C3H8(v)+M=C3H8+MN2(c3pu)+C4H10=>N2+C4H8+2H(T)N2(+)+C2H4=>N2+C2H3(+)+HO(-)+NO2(+) =>NO2+ON2(c3pu)+C4H10=>N2+C4H8+H2 LT /0.0 0.0/ HO2(v) = O2 + HO(-)+NO2(+) =>NO+O+ON2(c3pu)+C4H10=>N2+CH3+C3H7 H2O2(v) = OH + OHN2(c3pu)+C4H10=>N2+C2H5+C2H5 C2H3(+)+e=C2H2+HO(-)+NO3(+) =>NO+O+O2C4H10(v)+M=C4H10+MO(-)+N2O(+) =>N+NO+OLT /0.0 0.0/ N2(c3pu)+C5H12=>N2+C5H11+H(T)N2(+)+C2H6=>N2+C2H5(+)+H!H2O2+O = OH+HO2O(-)+N2O(+) =>N2O+ON2(c3pu)+C5H12=>N2+C5H10+2H(T)N2(+)+C3H8=>N2+C3H7(+)+HO(-)+N3O(+) =>N2+NO+O!H2O2+OH = H2O+HO2C5H12(v)+M=C5H12+MN2(c3pu)+C5H12=>N2+C5H10+H2 N2(+)+C4H10=>N2+C4H9(+)+HO(-)+N2O2(+)=>NO+NO+ON2(c3pu)+C5H12=>N2+CH3+C4H9 LT /0.0 0.0/ !HO2+0 OH+O2 N2(+)+C5H12=>N2+C5H11(+)+HO(-)+N2O2(+)=>N2+O+O2N2(c3pu)+C5H12=>N2+C2H5+C3H7 N2 + CO2(v) = CO2 + N2(v)!HO2+OH = H2O+O2

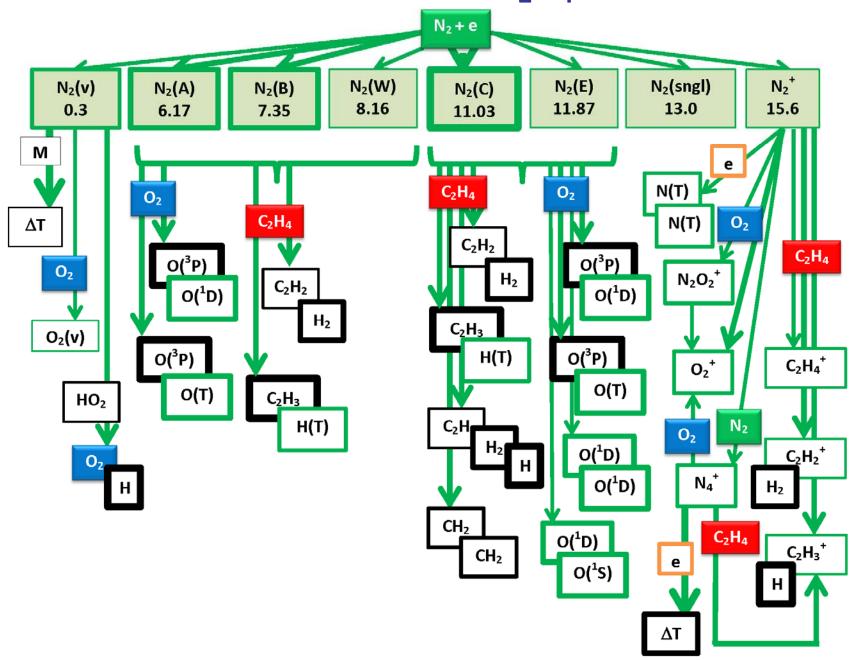
#### PAC Pathways: C<sub>2</sub>H<sub>4</sub>-air



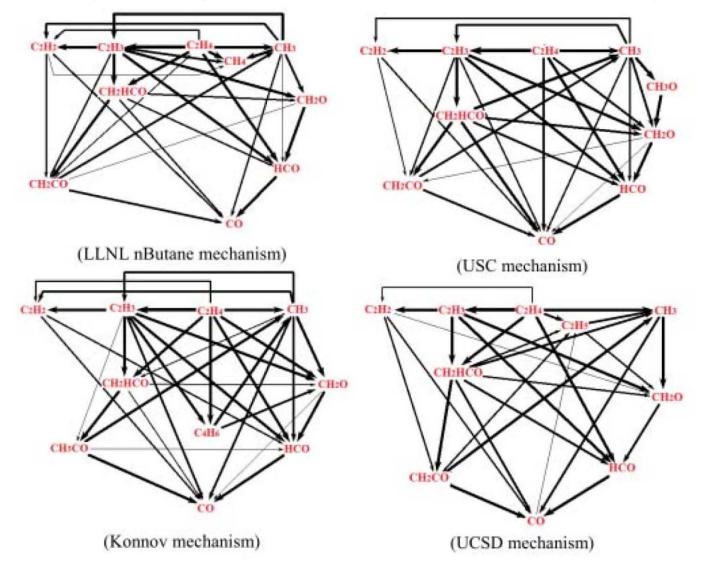
### PAC Pathways: C<sub>2</sub>H<sub>4</sub>-air



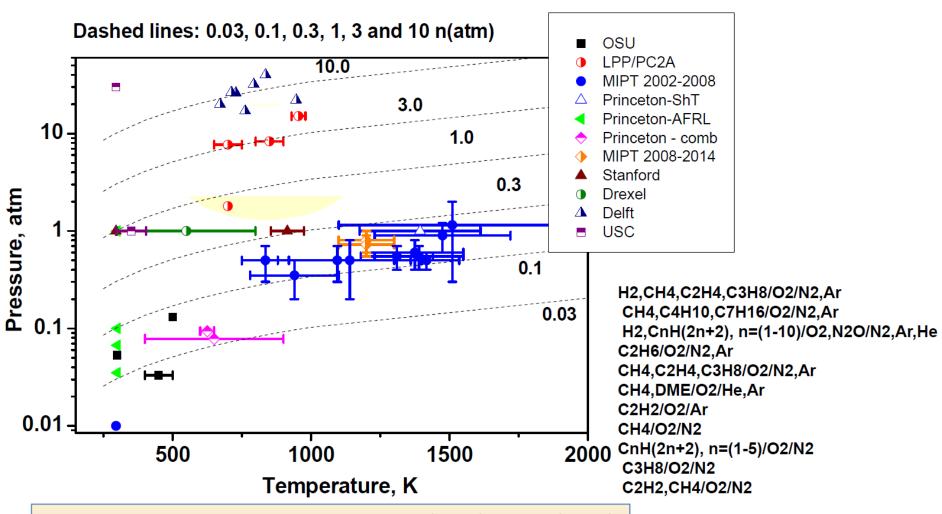
#### PAC Pathways: C<sub>2</sub>H<sub>4</sub>-air



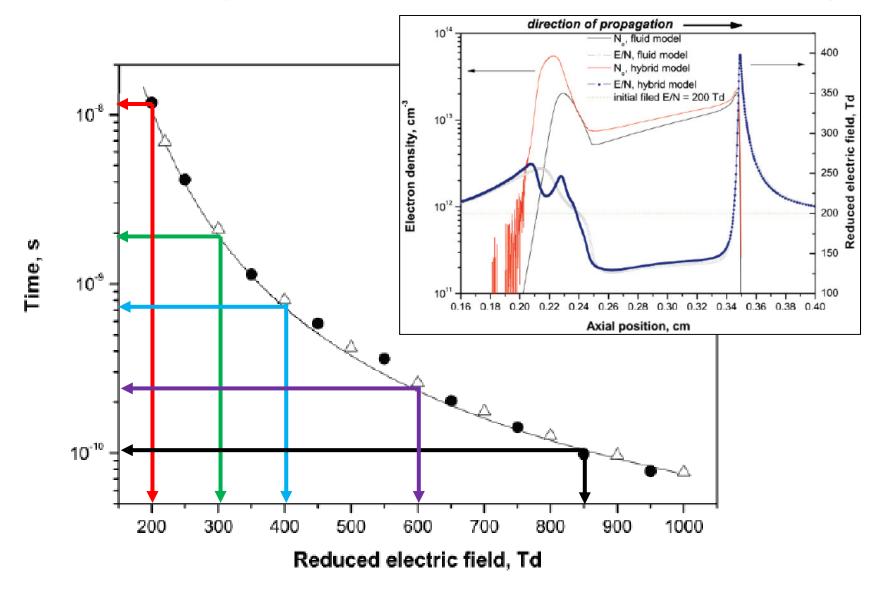
Pathway analysis in ignition process at the time of T=1360K for  $\emptyset=1$ , Ar=92%, P=2.1atm and initial T=1350K. Konnov, 2014



#### Where PAC Experimental Data is Available



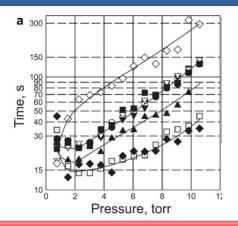
## Avalanche to Streamer Transition in Uniform Electric Field (air, 1 bar, 300 K, 1 cm, various E/n)



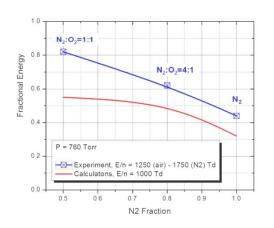
#### **Mechanism Validation**

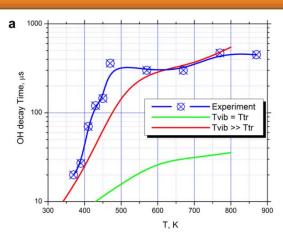
T = 300 K T = 300 - 800 K T = 800 - 1700 K

#### Slow Oxidation of H<sub>2</sub>, C1-C10 P = 1-10 Torr

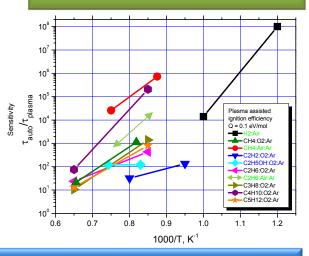


Fast Gas Heating Mechanism.  $N_2$ - $O_2$  mixtures P = 0.2 - 1 atm

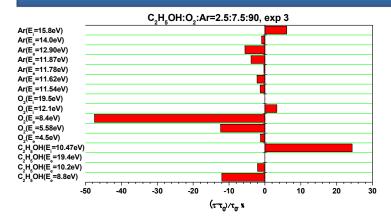


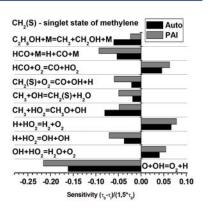


Oxidation Chains Development in Lean H<sub>2</sub>, CO, C1-C4 - Air Mixtures. P = 1 atm Ignition Delay Time Reduction.  $H_2$ , C1-C5,  $C_2H_2$ ,  $C_2H_5OH - O_2$ -Ar Mixtures. P = 0.3-0.5 atm

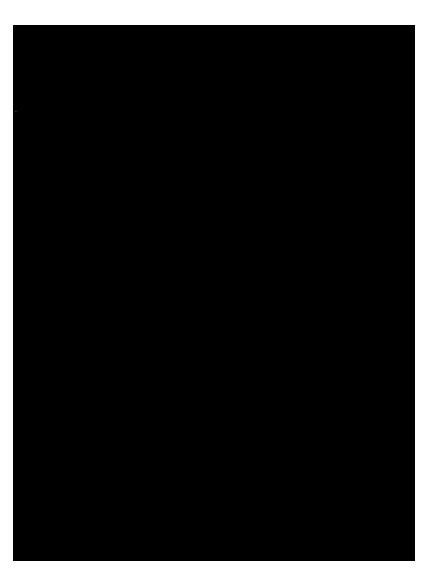


#### **Sensitivity Analysis for Discharge and Combustion Stages**





### **SDBD** Discharge and Fast Heating



Gate = 0.5 ns

Time shift between frames is 1 ns

The movie duration is 41 ns

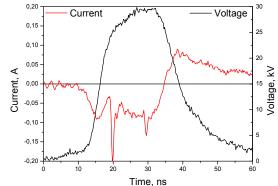
*Impulse Parameters* 

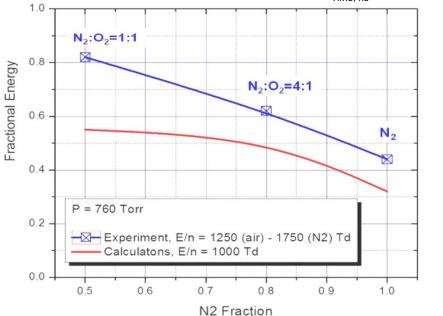
V = 14 kV

 $t_{1/2} = 20 \text{ ns}$ 

Frequency = 1 kHz

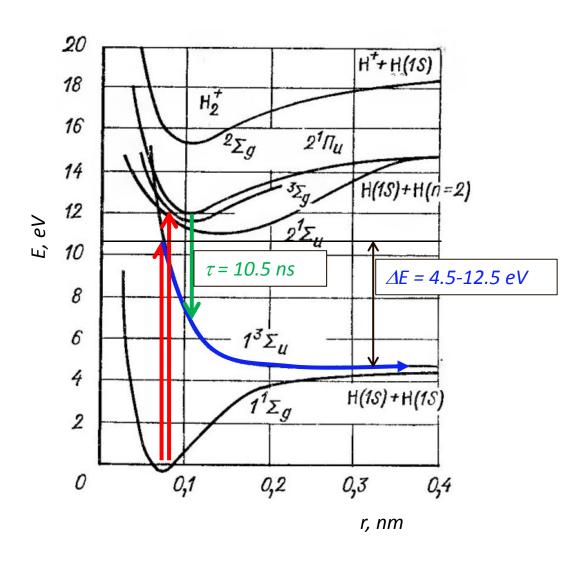
Velocity = 0.4 mm/ns





Physics of Nonequilibrium Systems Laboratory

## Potential Energy Curves of Molecular Hydrogen

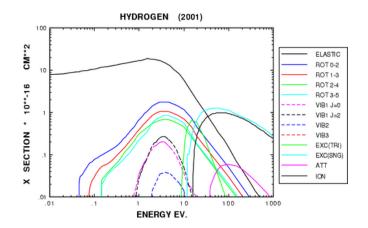


$$H_2(b^3\Sigma_u)$$
, 8.9 eV  
 $\sigma_{max} = 0.33 A^2 (17 eV)$ 

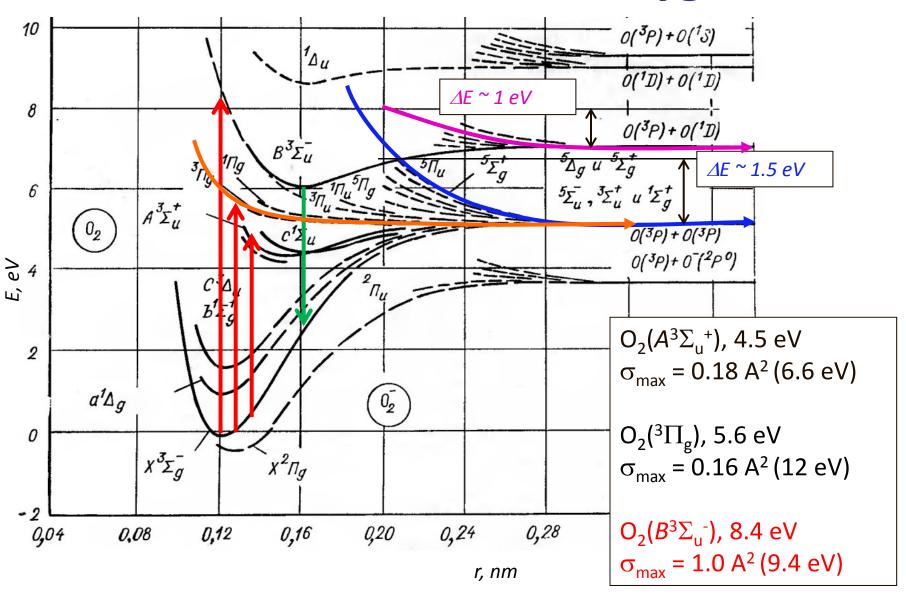
$$H_2(a^3\Sigma_g)$$
, 11.8 eV  $\sigma_{max} = 0.12 A^2 (15 eV)$ 

$$H_2(B^1\Sigma_u)$$
, 11.3 eV  
 $\sigma_{max} = 0.48 A^2 (40 eV)$ 

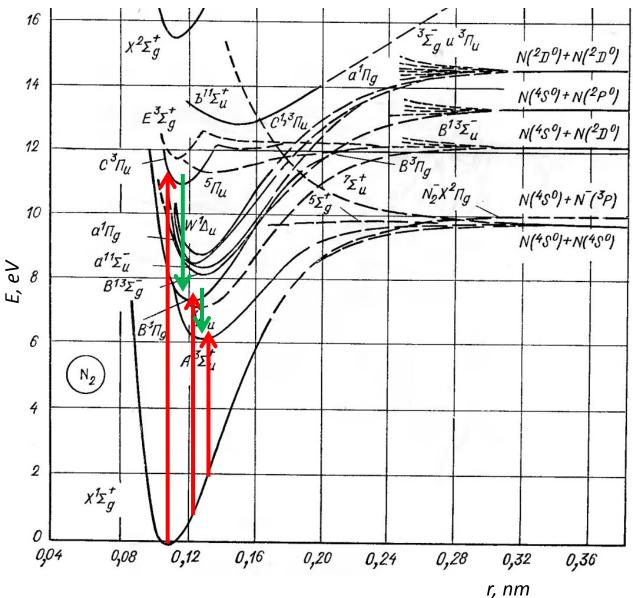
$$H_2(C^1\Pi_u)$$
, 12.4 eV  
 $\sigma_{max} = 0.40 A^2 (40 eV)$ 



## Potential Energy Curves of Molecular Oxygen



## Potential Energy Curves of Molecular Nitrogen



$$N_2(A^3\Sigma_u^+)$$
, 6.2 eV  $\sigma_{max} = 0.08 A^2 (10 eV)$ 

$$N_2(B^3\Pi_g)$$
, 7.35 eV  
 $\sigma_{max} = 0.20 A^2 (12 eV)$ 

$$N_2(C^3\Pi_u)$$
, 11.03 eV  $\sigma_{max} = 0.98 A^2 (14 eV)$ 

## Major Channels of Hot Atoms Production

$$N_2 + e = N_2(C^3\Pi_{II}) + e;$$
  $k = f(E/n)$ 

$$N_2(C^3\Pi_{11}) + H_2 = N_2 + 2H(^1S) + 6.55 \text{ eV}; \quad k = 3.2 \times 10^{-10} \text{ cm}^3/\text{s}$$

$$N_2(C^3\Pi_{II}) + O_2 = N_2 + 2O(^3P_1^1D) + 3.9 \text{ eV}; k = 2.7x10^{-10} \text{ cm}^3/\text{s}$$

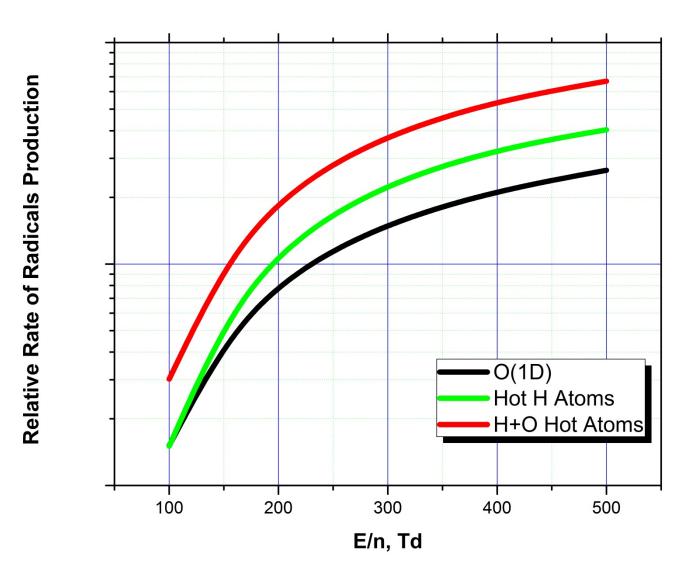
$$O_2 + e = e + 2O(^3P,^1D) + 1.3 eV;$$
  $k = f(E/n)$ 

$$H_2 + e = e + 2H(^1S) + 4.4 eV;$$
  $k = f(E/n)$ 

### **Chain Initiation/Branching Reactions**

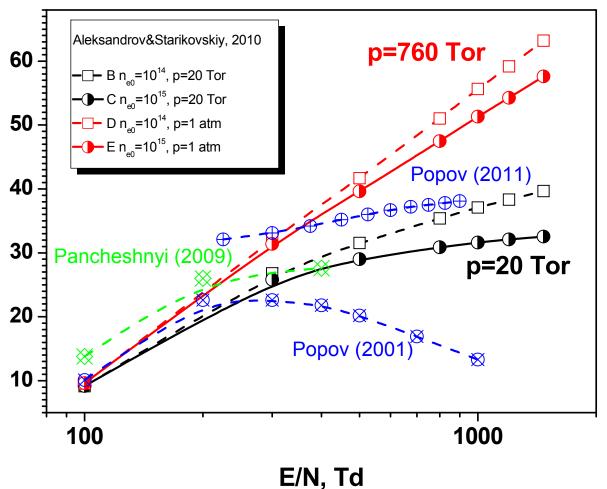
```
H + O_2 = O + OH
                                                          k = 1.6x10^{-10} x exp(-7470/T) cm^3/s
                                                          k(300) = 2.5 \times 10^{-21} \text{ cm}^3/\text{s}
                                                          k(hot) = 1.6x10^{-10} cm^3/s
                                                          k(300, 1 \text{ atm}) = 1.6 \times 10^{-12} \text{ cm}^3/\text{s} T_{crit} \sim T_{autoignition}
H + O_2 + M = HO_2 + M
O + H_2 = H + OH
                                                          k = 8.5 \times 10^{-20} \times T^{2.67} \times exp(-3160/T) cm^3/s
                                                          k(300) = 9.3x10^{-18} \text{ cm}^3/\text{s}
                                                          k(hot) = 1.5x10^{-10} cm^3/s
                                                          k(^{1}D) = 1.1x10^{-10} \text{ cm}^{3}/\text{s}
O + O_2 + M = O_2 + M
                                                          k(300, 1 atm) = 2.2x10^{-14} cm^3/s
                                                                                                                          T<sub>crit</sub> ~ 650K
                                                          k \sim 2m/M k_{gk} \sim 1.6x10^{-10} cm^3/s
H(hot) + (N_2, H_2) = H + (N_2, H_2)
                                                          k \sim 2m/M k_{gk} \sim 1.3 \times 10^{-10} \text{ cm}^3/\text{s}
O(hot) + (N_2, O_2) = O + (N_2, O_2)
H(hot) + O_2 = H + O + O
H(hot) + H_2 = H + H + H
O(^{1}D) + (M) = O + (M)
                                                          k = 2.6 \times 10^{-11} \text{ cm}^3/\text{s} \text{ (M = O_2)}
                                                          k = 1.3x10^{-11} \text{ cm}^3/\text{s} \text{ (M = N<sub>2</sub>)}
                                                          k = 5.2 \times 10^{-11} \text{ cm}^3/\text{s} \text{ (M = H<sub>2</sub>)}
```

## Radicals Production Increase in Cold H<sub>2</sub>-Air Mixture Due to Hot Atoms Formation



## Mechanism of Fast Heating in Discharge Plasmas (high E/N)

 $e + O_2^+ \rightarrow O + O^* + \Delta E$   $O_2^- + O_2^+ + M \rightarrow 2O_2 + M + \Delta E$   $e + O_4^+ \rightarrow O_2 + O_2 + \Delta E$ 

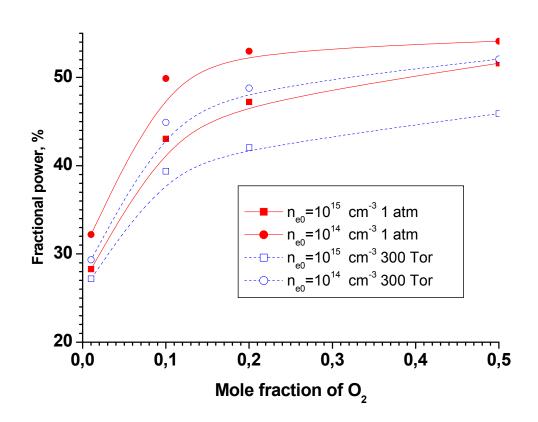


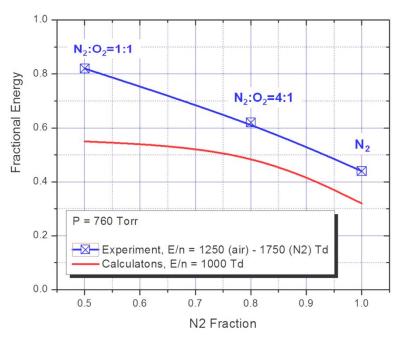
#### High (> 200 Td) E/N:

electron-ion and ion-ion recombination kinetics

## Fractional Electron Power Transferred Into Heat in N<sub>2</sub>:O<sub>2</sub> Mixtures

$$E/N = 10^3 \text{ Td}$$



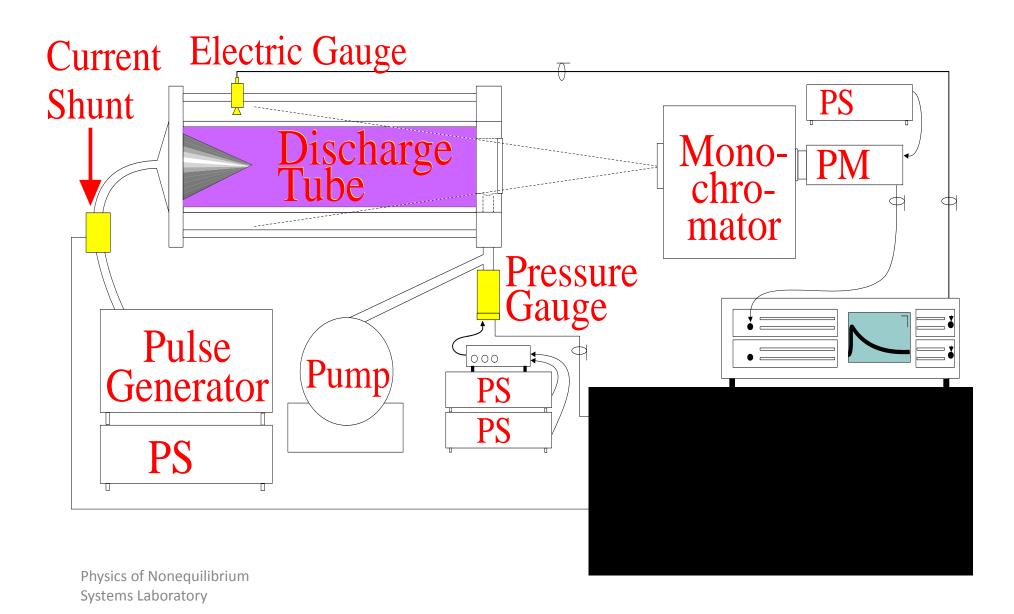


Oxygen is required for efficient fast heating!

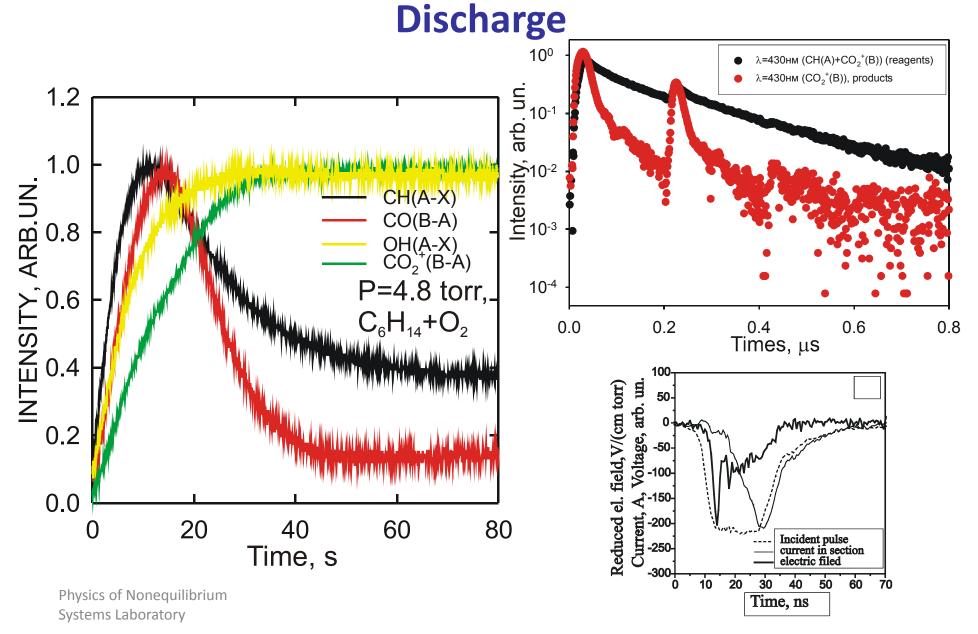
$$AB^+ + O_2^- (+ M) \rightarrow products + \Delta E$$

$$N_2(A,B,C,a) + O_2 \rightarrow e + 2O + \Delta E$$

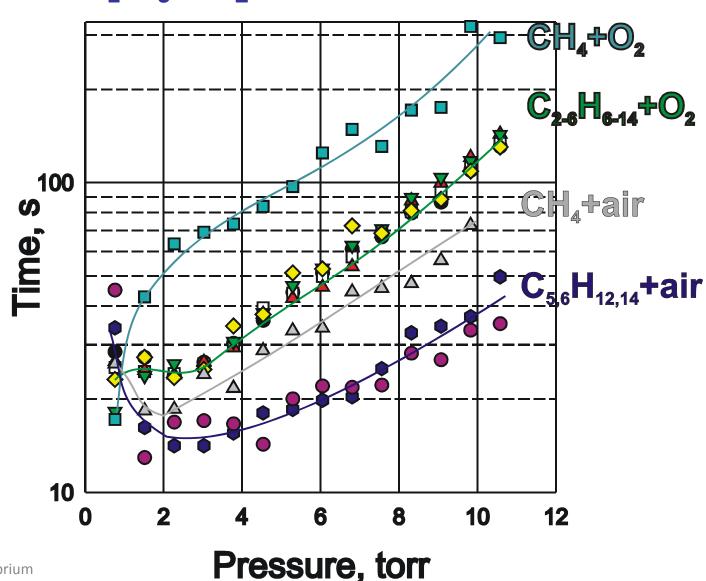
### **Experimental Setup**



Hexane Oxidation by Pulsed Nanosecond

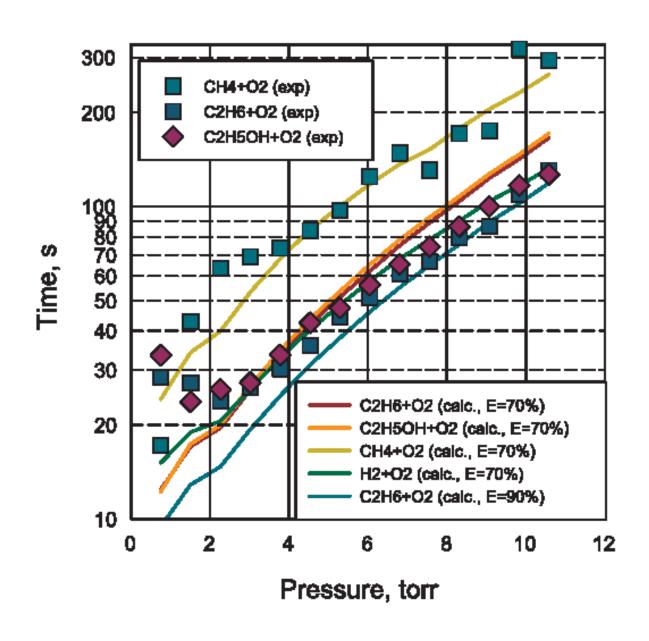


## Hydrocarbon Oxidation Efficiency for $C_1$ - $C_6$ / $O_2$ / Air Mixtures



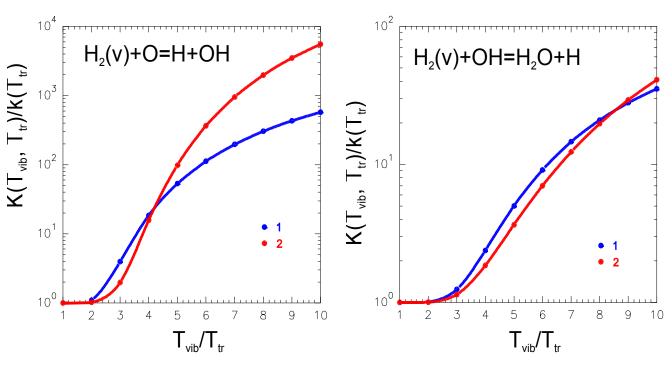
Physics of Nonequilibrium Systems Laboratory

#### **Calculated and Measured Times of Oxidation**



## Chemical Reactions with Excited Reagents

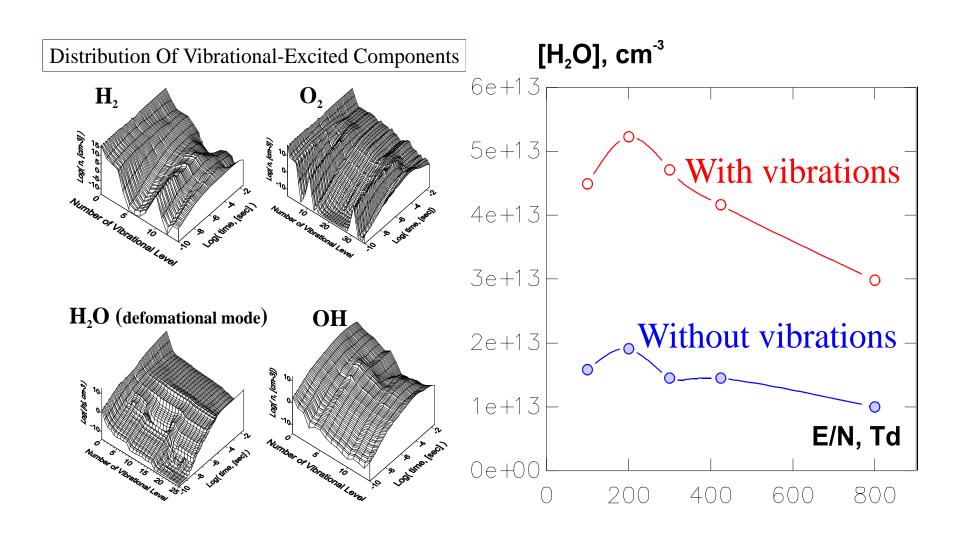
AB(v)+C = A + BC(w)Rate constant from modified  $\alpha$ -model (Starikovskii, Lashin 1996)



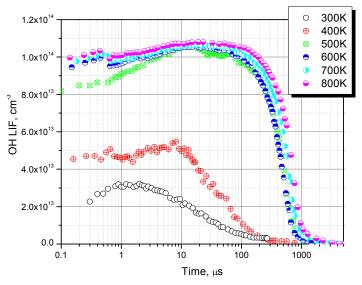
$$H_2(v=1) + O = OH(w=1) + H$$
 (R1)  
 $H_2(v=0) + O = OH(w=0) + H$  (R2)

 $(k_{R1}/k_{R2})_{exp} = 2600 (O'Neal, Benson 1973); (k_{R1}/k_{R2})_{theor} = 2750$ 

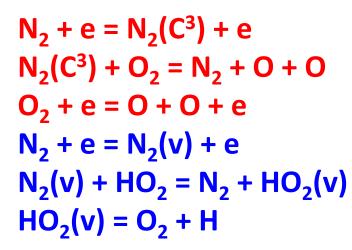
#### **Kinetics. Influence of Vibrations**

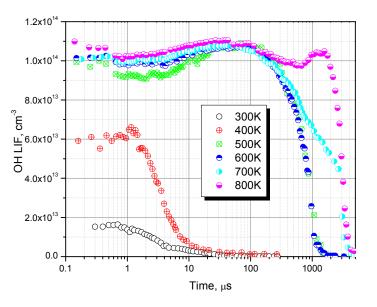


### Influence of Vibrational Excitation on Low-Temperature Kinetics

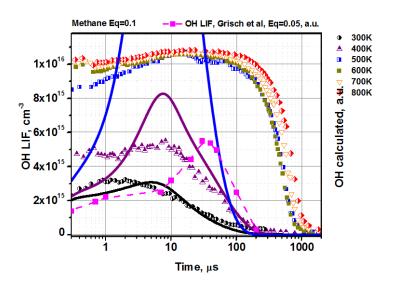


CH4-air





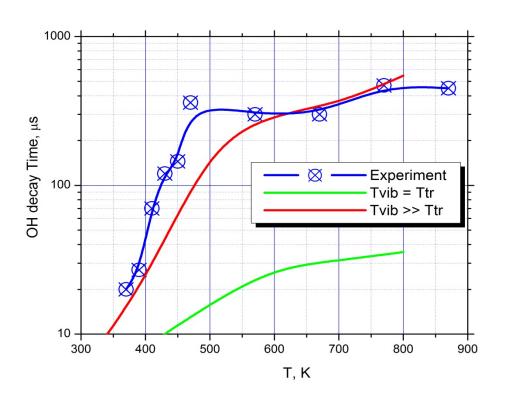
C4H10-air

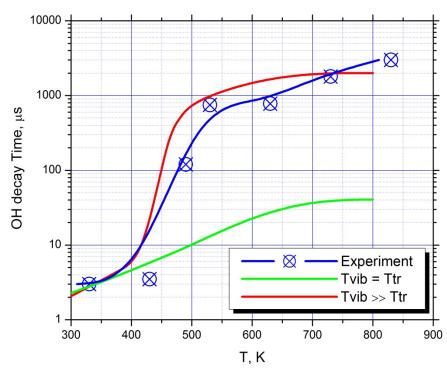


Experiments: L Wu, J Lane, N P Cernansky, D L Miller, A A Fridman, A Yu Starikovskiy, *Proc. of Comb. Inst.*, 2010

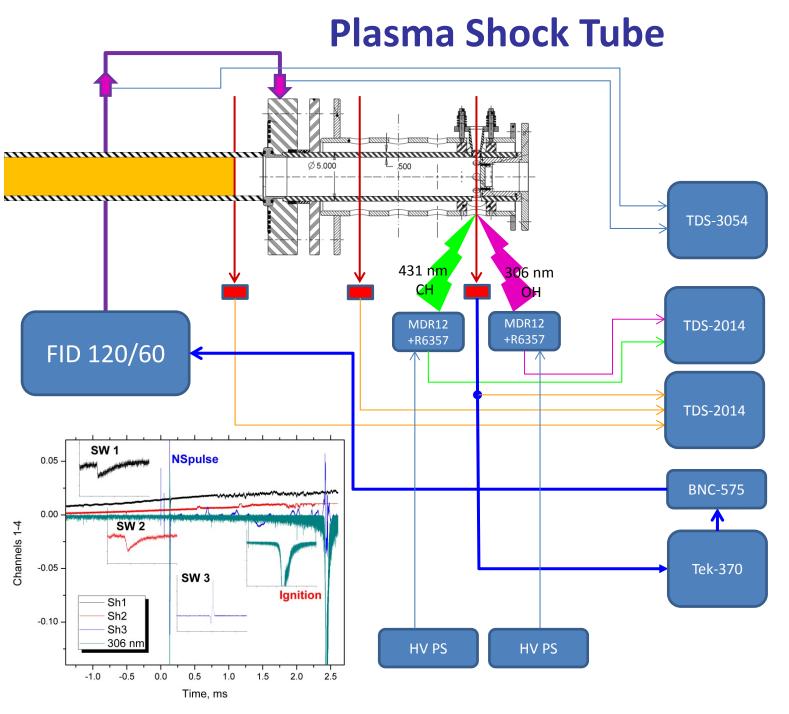
Modelling: D Levko, A I Schedrin, V V Naumov, S Starikovskaia, 2010

### Influence of Vibrational Excitation on Low-Temperature Kinetics: H<sub>2</sub>O<sub>2</sub> Decomposition



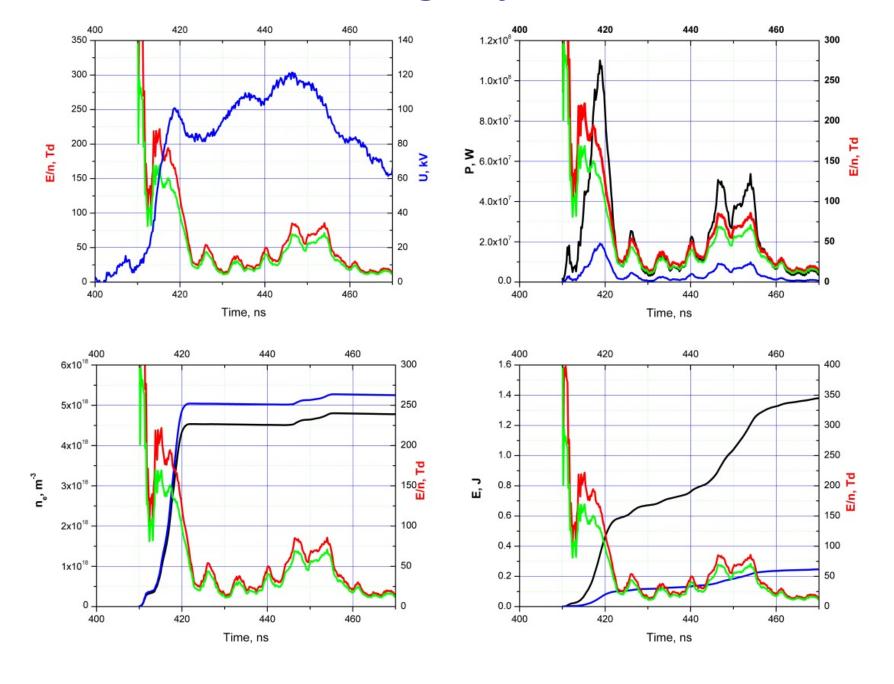


Measured and calculated OH decay time. P = 1 atm. a)  $3\%H_2 + air$ ; b)  $0.3\%C_4H_{10} + air$ .



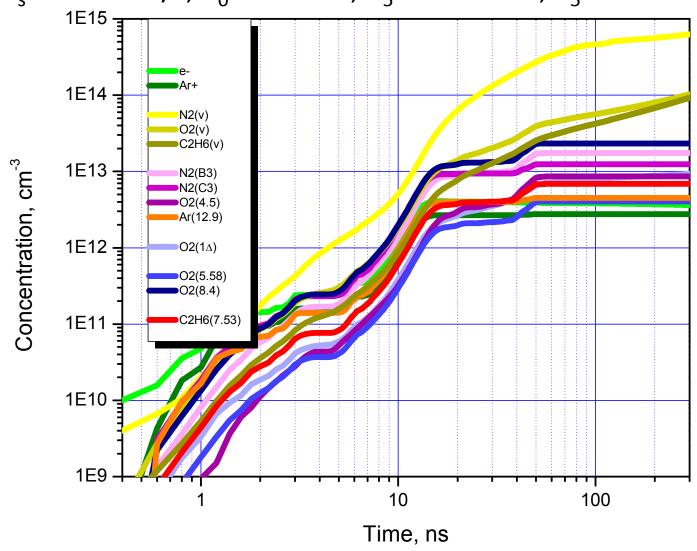


#### **Discharge Dynamics**

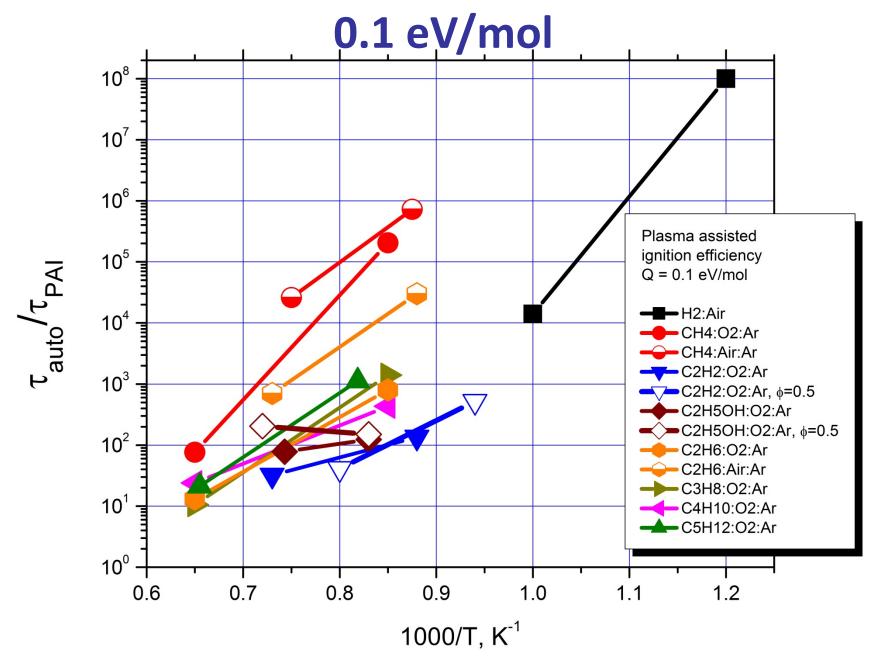


#### **Active Particle Production – Discharge Phase**

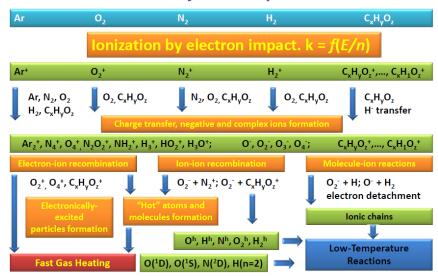
 $U_s = 943.6 \text{ m/s}$ ;  $P_0 = 17 \text{ Torr}$ ;  $P_5 = 1.04 \text{ atm}$ ;  $T_5 = 1525 \text{ K}$ 



### **Plasma Ignition Sensitivity**

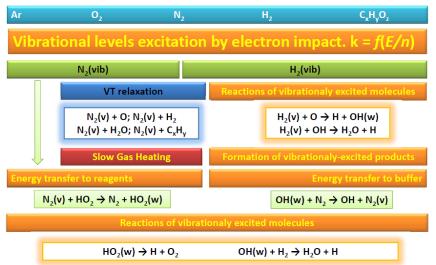


**Major Pathways** 



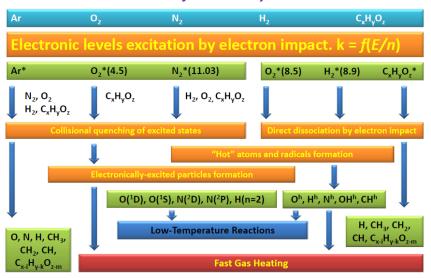
#### **Princeton Plasma Combustion Kinetics**

**Major Pathways** 



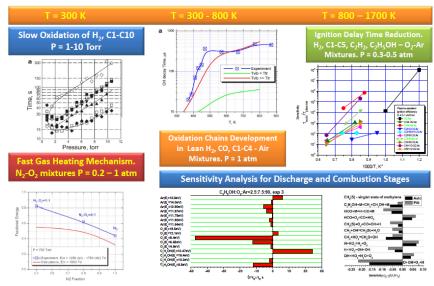
#### **Princeton Plasma Combustion Kinetics**

**Major Pathways** 

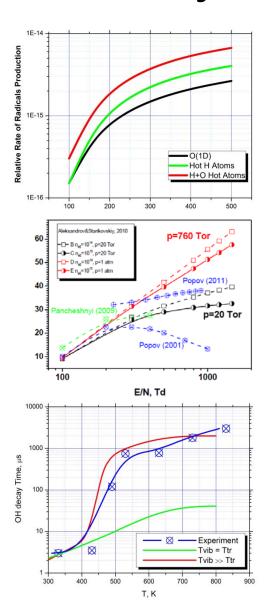


#### **Princeton Plasma Combustion Kinetics**

**Mechanism Validation** 



#### **Other Major Results**

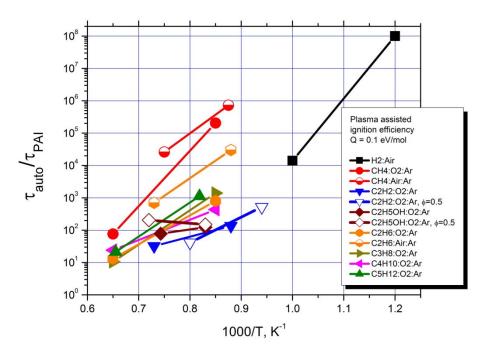


#### **3 NEW MECHANISMS:**

- Radicals Production Increase Due to Translationally Hot Atoms Formation
- Mechanism of Fast Heating in Plasmas at high E/n
- Vibrational Decomposition of Peroxides (HO2, H2O2, etc)

#### **EXPERIMENTAL DATABASE:**

 Plasma Ignition Delay Time database for H2, C1-C5, acetylene, ethylene, ethanol



### The work was supported by

# AFOSR Technical Monitor Dr Chiping Li

